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REEVALUATION OF BACKGROUND IODINE-129 CONCENTRATIONS IN WATER FROM THE SNAKE RIVER PLAIN AQUIFER, IDAHO, 2003

U.S. GEOLOGICAL SURVEY WATER-RESOURCES INVESTIGATIONS REPORT 03-4106



Prepared in cooperation with the U.S. DEPARTMENT OF ENERGY

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U.S. GEOLOGICAL SURVEY

Water-Resources Investigations Report 03-4106

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**Idaho Falls, Idaho
May 2003**

U.S. DEPARTMENT OF THE INTERIOR
GALE A. NORTON, Secretary

U.S. GEOLOGICAL SURVEY
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CONVERSION FACTORS AND OTHER ABBREVIATED UNITS

<u>Multiply</u>	<u>By</u>	<u>To Obtain</u>
centimeter per year (cm/yr)	0.3937	inch per year (in/yr)
cubic meter (m ³)	35.31	cubic foot (ft ³)
curie (Ci)	3.7 x 10 ¹⁰	becquerel (Bq)
attocurie per liter (aCi/L)	3.7 x 10 ⁻⁸	becquerel per liter (Bq/L)
gram per cubic centimeter (g/cm ³)	0.5780	ounce per cubic inch (oz/in ³)
kilometer (km)	0.6214	mile (mi)
liter (L)	0.2642	gallon (gal)
meter (m)	3.281	foot (ft)
meter per kilometer (m/km)	5.280	foot per mile (ft/mi)
square kilometer (km ²)	0.3861	square mile (mi ²)
square meter per day (m ² /d)	10.76	square foot per day (ft ² /day)
degree Celsius (°C)	[C (9/5) + 32]	degree Fahrenheit (°F)
atoms/L iodine-129	(8.4 x 10 ⁻¹⁴ min ⁻¹)/2.22	picocuries/L iodine-129

Other abbreviated units used in this report:

atoms/cm² (atoms per square centimeter)
atoms/g (atoms per gram)
atoms/L (atoms per liter)
aCi/g (attocuries per gram)
m³/s (cubic meters per second)
μg/L (micrograms per liter)
mg/L (milligrams per liter)
pCi/L (picocuries per liter)
ppm (parts per million)

REEVALUATION OF BACKGROUND IODINE-129 CONCENTRATIONS IN WATER FROM THE EASTERN SNAKE RIVER PLAIN AQUIFER, IDAHO, 2003

By L. DeWayne Cecil, L. Flint Hall, and Jaromy R. Green

Abstract

Background concentrations of iodine-129 (^{129}I , half-life = 15.7 million years) resulting from natural production in the earth's atmosphere, *in situ* production in the earth by spontaneous fission of uranium-238 (^{238}U), and fallout from nuclear-weapons tests conducted in the 1950s and 1960s were reevaluated on the basis of 52 analyses of ground- and surface-water samples collected from the eastern Snake River Plain in southeastern Idaho. The background concentration estimated using the results of a subset of 30 ground-water samples analyzed in this reevaluation is 5.4 attocuries per liter (aCi/L; 1 aCi = 10^{-18} curies) and the 95-percent nonparametric confidence interval is 5.2 to 10.0 aCi/L. In a previous study, a background ^{129}I concentration was estimated on the basis of analyses of water samples from 16 sites on or tributary to the eastern Snake River Plain. At the 99-percent confidence level, background concentrations of ^{129}I in that study were less than or equal to 8.2 aCi/L.

During 1993–94, 34 water samples from 32 additional sites were analyzed for ^{129}I to better establish the background concentrations in surface and ground water from the eastern Snake River Plain that is presumed to be unaffected by waste-disposal practices at the Idaho National Engineering and Environmental Laboratory (INEEL). Surface water contained larger ^{129}I concentrations than water from springs and wells contained. Because surface water is more likely to be affected by anthropogenic fallout and evapotranspiration, background ^{129}I concentrations were estimated in the current research using the laboratory results of ground-water samples that were assumed to be unaffected by INEEL disposal practices.

INTRODUCTION

Background concentrations of iodine-129 (^{129}I) in water from the eastern Snake River Plain (SRP) aquifer need to be established to evaluate the long-term effects of past practices for ^{129}I disposal in wastewater at the Idaho National Engineering and Environmental Laboratory (INEEL, fig. 1). Until 1993, spent nuclear-fuel elements from government-owned reactors were processed at the Idaho Nuclear Technology and Engineering Center (INTEC, fig. 1) at the INEEL to recover uranium (U). Between 1953 and 1990, wastewater discharged to injection wells and infiltration ponds at the INTEC contained an estimated 0.56 to 1.18 curies (Ci) of ^{129}I (Mann and Beasley, 1994a).

Monitoring for ^{129}I began in 1976 for wastewater and in 1977 for ground water (Barracough and others, 1982). Work by the U.S. Geological Survey (USGS) on the INEEL from 1977 through 1988 identified ^{129}I in water from wells at the INEEL and demonstrated that the isotope was present in measurable concentrations and had traveled with water in the aquifer. With the availability of an accelerator mass spectroscopy-based (AMS) method for ^{129}I measurements providing a much lower level of detection, background levels of the isotope could be determined. In 1992, 16 water samples from selected sites on or tributary to the eastern SRP (table 1 and fig. 1) were analyzed to determine background concentrations of ^{129}I (Mann and Beasley, 1994a). For this report, background ^{129}I concentrations are defined as those levels in water resulting from natural production, as well as fallout from atmospheric weapons tests in the 1950s and 1960s. The present study strengthens the foundation laid by previous work by reporting ^{129}I analyses for 34 additional water

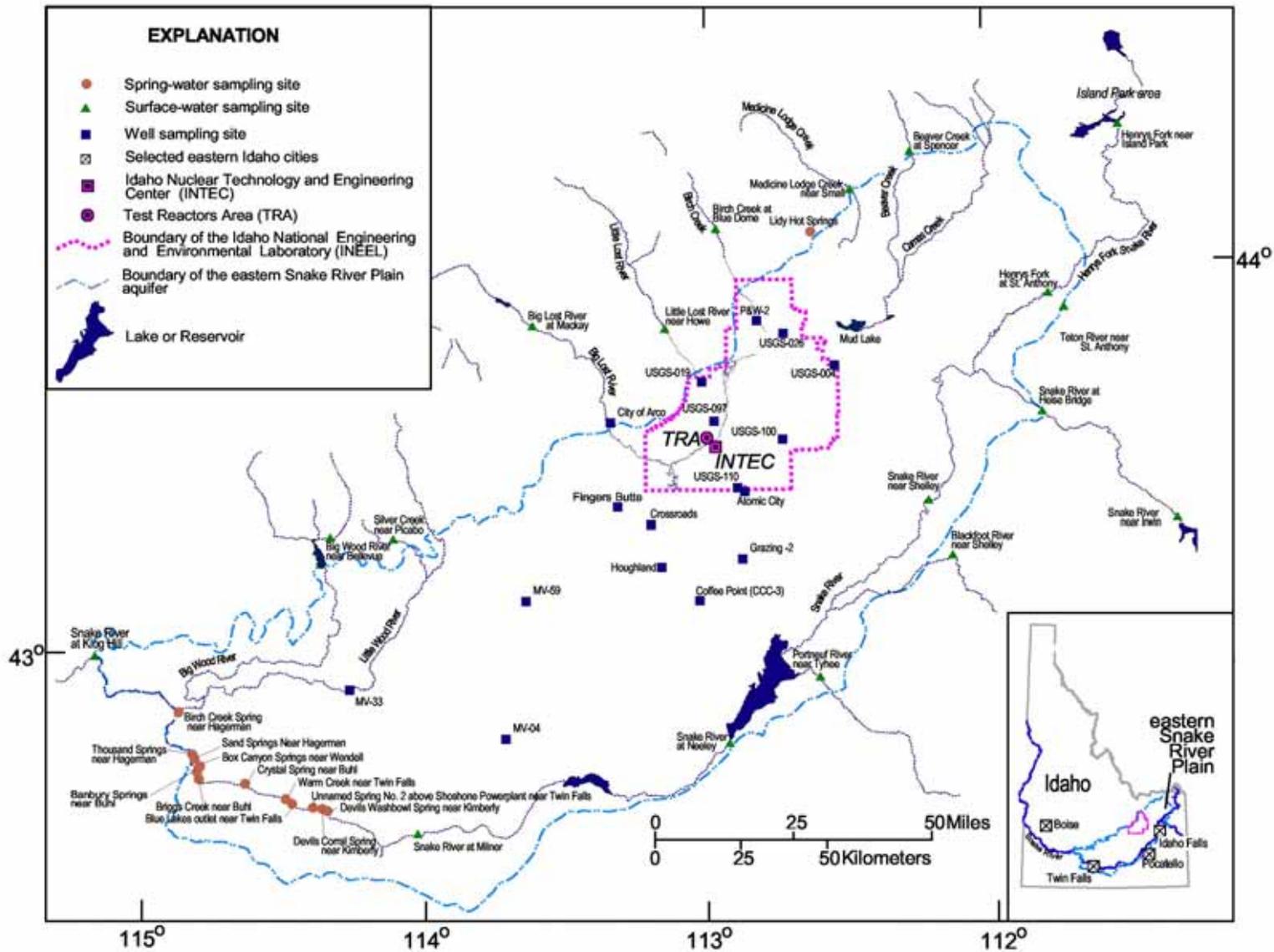


Figure 1. Map showing location of sampling sites, eastern Snake River Plain, Idaho.

samples from 32 sites on the SRP to reevaluate background concentrations (tables 2 and 3). These 34 water samples were collected during 1993–94 and included water samples selected from USGS archives (1992–93) and two sites sampled in both 1992 and 1994. Additionally, blind replicate samples were collected from two locations and analyzed for ^{129}I as a quality assurance/quality control (QA/QC) measure.

This study was conducted by the USGS, the State of Idaho's INEEL Oversight Program, the U.S. Department of Energy (DOE), U.S. Environmental Protection Agency's (EPA) Region 10, and the Idaho Department of Health and Welfare, Division of Environmental Quality.

Purpose and Scope

This report describes analytical results for 34 water samples collected from 32 sites to reevaluate background concentrations of ^{129}I in the eastern SRP aquifer. Some samples were collected during 1993–94 and others were selected from the USGS archive sample library at the INEEL. In addition, data analysis in this report includes 16 analytical results for samples collected in 1992 and reported by Mann and Beasley (1994a).

Description of the Study Area

The eastern SRP (fig. 1) is a structural down-warp filled with basalt that is generally within 3 m of the land surface (Whitehead, 1992). Unconsolidated sediments overlie the margins of the plain and are interbedded with basalt and sediments at depth. The basalt is several hundred to as much as 1,500 m thick and underlies most of the plain. Fractures and vesicular zones near the surface of the basalt flows may be highly transmissive of ground water. Reported transmissivities for the eastern SRP aquifer range from 0.1 to more than 70,000 m^2/d , a range of nearly six orders of magnitude (Ackerman, 1991). Depth to ground water in the basalt aquifer at the INEEL varies from 60 m below land surface in the northern part to more than 275 m in the southern part. The hydraulic gradient is about 1 m/km and horizontal flow velocities range from 1 to 6 m/d . This range is based on the distribution of chlorine-36 (^{36}Cl) through time

as determined from analysis of archived samples (Cecil, 2000) and equates to a travel time of 100 to 200 years for water to travel from beneath the INEEL to springs that discharge at the terminus of the eastern SRP aquifer, and about 300 years or more for water to travel from all places within the aquifer to the terminus of the eastern SRP aquifer (Ackerman, 1995).

Long-term (1950–88) average precipitation in the vicinity of the INEEL is 22 cm/yr (Clawson and others, 1989). About 40 percent of the long-term precipitation on the eastern SRP is rainfall between April and September. However, as a result of evapotranspiration, generally less than 5 percent of that precipitation infiltrates the surface locally on the SRP (Cecil and others, 1992). Recharge to the SRP aquifer is from precipitation in the mountains to the west, north, and east and from irrigation return flow and surface water. Five watersheds that recharge the aquifer are the Big Lost River, Little Lost River, Birch Creek, Camas Creek/Mud Lake, and the Snake River drainage (fig. 1).

The INEEL comprises about 2,300 km^2 of the eastern SRP. The INEEL was established in 1949 and is used by the DOE to construct and test nuclear reactors and to participate in various defense programs. Radiochemical and chemical wastes generated at the INEEL and other DOE facilities have been buried at the site since 1952. Additionally, from 1952 to 1984, low-level radioactive and chemical wastes were discharged into the SRP aquifer at the INTEC through a 182-m-deep disposal well. Since 1984 at the INTEC, and from 1952 to 1993 at the Test Reactors Area (fig. 1), these wastes also have been discharged to infiltration ponds.

Previous Investigations

Previous investigations of ^{129}I in water from the eastern SRP at the INEEL include those by Barraclough and others (1982), Lewis and Jensen (1985), Mann and others (1988), and Mann and Beasley (1994a, 1994b). Barraclough and others (1982) and Lewis and Jensen (1985) discussed results of sampling for ^{129}I at 20 sites in 1977 and 32 sites in 1981, respectively. Analytical results for samples collected in 1977 included concentra-

tions as large as 27 ± 1 pCi/L ($1 \text{ pCi} = 10^{-12} \text{ Ci}$); concentrations were detectable (about 0.9 pCi/L) as far as 5 km from the point of disposal at the INTEC. Concentrations as large as 41 ± 2 pCi/L and detectable as far as 10 km from the point of disposal were reported for the 1981 sampling (Lewis and Jensen, 1985). The major difference between the 1977 and 1981 results was the reduction in the reporting level from approximately 0.9 to 0.05 pCi/L. The increase in analytical sensitivity was a result of an increase in sample size used for the neutron-activation analysis method from 1 to 4 L. Mann and others (1988) again analyzed water samples for ^{129}I by neutron activation, collecting from sites visited in 1977 and 1981, as well as from additional sites. Although the portion of the aquifer where concentrations exceeded the reporting level had changed little from 1981 to 1986, the maximum concentration had decreased by an order of magnitude to 3.6 ± 0.4 pCi/L from the largest concentration reported in 1981.

During 1990–91, Mann and Beasley (1994b) collected samples from 51 wells at and near the INEEL and utilized AMS analysis methods for increased sensitivity (two to six times more sensitive than neutron activation). Included were sites sampled in 1977, 1981, and 1986. Although the maximum concentration measured during 1990–91 (3.81 ± 0.19 pCi/L) was indistinguishable from the maximum concentration measured in 1986 (3.6 ± 0.4 pCi/L), the average concentration for sites sampled previously decreased, continuing the downward trend observed for the 1986 sampling. Sampling during 1990–91 included one site upgradient from the point of disposal at the INTEC. The result from this site established a preliminary background concentration of 0.9 ± 0.2 aCi/L. Also included were two wells 6 and 13 km south of the INEEL southern boundary. Water from these wells contained concentrations of 10 ± 1 and 30 ± 2 aCi/L, which documented migration of ^{129}I as much as 26 km south of the point of disposal and 13 km beyond the INEEL boundary.

In 1992, Mann and Beasley (1994a) sampled 16 sites not likely to have been impacted by INEEL waste disposal to better define background levels of ^{129}I . Concentrations ranged from 0.1 ± 0.1 to 8.1 ± 0.6 aCi/L and averaged 3.3 ± 2.1 aCi/L. At

the 99-percent confidence level, background concentrations of ^{129}I were estimated to be less than or equal to 8.2 aCi/L. For comparison, the Maximum Contaminant Level (MCL) established by the EPA for ^{129}I in drinking water is 1 pCi/L (U.S. Environmental Protection Agency, 1991), or more than five orders of magnitude larger than the estimated background concentration of 8.2 aCi/L.

SOURCES OF IODINE-129 IN THE EASTERN SNAKE RIVER PLAIN AQUIFER

The sources of ^{129}I in the ground and surface water of the eastern SRP are (1) natural atmospheric production and *in situ* spontaneous fission of ^{238}U in the lithosphere (Fabryka-Martin and others, 1985), (2) above-ground testing of nuclear devices, and (3) releases from nuclear-power and nuclear-fuel reprocessing facilities (National Council on Radiation Protection and Measurements, 1983). These sources are reviewed in this report to determine their relative importance as contributors of ^{129}I to SRP water and to provide a basis for determining their relation to measured ^{129}I concentrations.

Natural Sources

The principal natural sources of ^{129}I to water of the SRP are (1) production in the atmosphere by cosmic-ray spallation of xenon isotopes, and (2) production from the spontaneous fission of ^{238}U isotopes *in situ* in the lithosphere. Global model calculations by Fabryka-Martin and others (1985) indicate an equilibrium ^{129}I areal inventory in surface soils to 1-m in depth to be about 2×10^5 atoms/cm². On the basis of data from 30-cm-deep soil cores collected worldwide by the DOE's Environmental Measurements Laboratory (EML) (Beasley and others, 1998), average soil density is 1.3 g/cm³ and the natural ^{129}I concentration in surface soil is on the order of 1.5×10^3 atoms/g of soil. Because of the biophilic nature of iodine, which results in sequestration in organic materials (Wagner and others, 1996), and because most organic materials in soils are in the upper few centimeters

of the soil column, ^{129}I concentration typically are on the order of 10^3 to 10^5 atoms/g in soils (Rao, 1997).

Natural ^{129}I concentrations in ground water have been measured across the globe. For example, concentrations from 2×10^{-3} to 1×10^{-1} aCi/L (5×10^4 to 2.5×10^6 atoms/L) were reported for ground water from the Great Artesian Basin, and from 1×10^{-2} to 4×10^{-1} aCi/L (2.5×10^5 to 9.8×10^6 atoms/L) for ground water from the Milk River aquifer in southern Saskatchewan, Canada (Fabryka-Martin and others, 1987). These background ^{129}I concentrations are less than that estimated in this report by one to three orders of magnitude. This suggests that water collected from the SRP aquifer system has a component of anthropogenically, as well as naturally, produced ^{129}I and that processes such as evapotranspiration probably have some effect on the concentrations of all isotopic tracers used to delineate contaminant plumes and to describe the hydrogeology at and near the INEEL (Cecil and others, 1999; Cecil, 2000).

Fabryka-Martin and others (1987) calculated the secular equilibrium ^{129}I concentration in rocks containing differing U concentrations and concluded that ^{129}I in sandstone and basalt (1 ppm U on average) reaches maximum concentrations of 1,500 atoms/g of rock at secular equilibrium. Shale (5 ppm U on average) and granite (3 ppm U on average) contained calculated concentrations of 6,800 and 4,200 atoms/g of rock, respectively. Given that age dates for SRP basalt have ranged from 2,000 to 5,000,000 years before present (Kuntz and others, 1992), ^{129}I concentrations in SRP basalt should be smaller than the concentrations reported by Fabryka-Martin and others (1987).

In contrast, if elevated concentrations of U exist in areas surrounding the plain, they would serve as additional sources of ^{129}I to surface and ground water. By analogy, reference is made to measurements of ^{129}I in the Alligator River Uranium Ore Province, Northern Territory, Australia, and in the Stripa granite formation in Sweden (Fabryka-Martin and others, 1987, 1988). In the Alligator River deposits (12,000 ppm U) the largest ground-water concentration of ^{129}I measured in the ore body was 0.5 aCi/L, although residence

times of the ground water pumped from the vicinity of the ore deposits were believed to be short, on the order of years to decades. Also, as a result of spatial variations in hydraulic conductivities, waters that had not contacted the ore deposits in the subsurface may have diluted the pumped ground water samples. In the case of the Stripa granite (which contained 44 ppm U, but with concentrations in fracture-lining minerals of as much as 5 percent U), water collected from 370 to 1,230 m below land surface contained progressively increasing ^{129}I concentrations ranging from 0.05 aCi/L (at 370 m) to 9.0 aCi/L (at 970 to 1,230 m). At the greater depths, the ^{129}I was ascribed entirely to *in situ* production. The residence times of water collected at 815 m (0.80 aCi/L) and that collected at 970 to 1,230 m were calculated to be 30,000 and 300,000 years, respectively, which would permit accumulation of ^{129}I in the water-bearing fracture zones.

For U to be a significant contributor of ^{129}I to water in wells, springs, and streams on and tributary to the eastern SRP, the U content of these bodies would need to equal or exceed that measured in the Alligator River ore deposits, and (or) water residence times in these host bodies would need to approximate (or exceed) those estimated for the Stripa granite. Even then, it is unlikely that the areal extent of the ore deposits would be sufficient to label enough water flowing through them to affect the nearly 4×10^{11} m³ of water estimated to be in the upper 170 m of the eastern SRP basalts (Lindholm, 1988). Additional factors that could influence the concentration of ^{129}I from *in situ* production include the distribution of the U in the basalt relative to the effective porosity of the basalt, the escape rate of ^{129}I into the flow system from the production sites within the aquifer matrix, and the water/rock ratio within the flow system. These factors were not determined for this study.

For analogy, Cecil and others (2000) calculated the *in situ* production of ^{36}Cl in the major water-bearing rock units of the eastern SRP aquifer system. The *in situ* production of ^{36}Cl was determined to be insignificant compared with concentrations measured in ground water near buried and injected nuclear waste sites at the INEEL. It

was concluded that this is a result of the relatively small concentration of U in the basalt of the SRP aquifer system and the relatively short residence time of water in this system. Therefore, because the residence time of the ground water is relatively short (300 years or less in most cases), compared with the half-life of ^{129}I (15.7 million years), concentrations of ^{129}I in ground water that result from *in situ* production by spontaneous fission of ^{238}U are expected to be insignificant.

Above-Ground Testing of Nuclear Devices

Haury and Schikarski (1977) estimated that approximately 12 Ci (3.2×10^{26} atoms) of ^{129}I were introduced into the atmosphere from above-ground testing of nuclear devices through 1976. If the deposition pattern of the ^{129}I followed that of plutonium, about 70 percent of the total atoms of ^{129}I would have been deposited in the Northern Hemisphere (Hardy and others, 1973). Of this 70 percent, 27 percent would have been deposited in the 40° to 50° north latitude band (area = 3.17×10^7 km²; Baumgartner and Reichel, 1975). Thus, the areal inventory of ^{129}I in soils of the SRP from global fallout would be about 2×10^8 atoms/cm². Gove and others (1994) have shown that, in southeastern Idaho soils, 90 percent of anthropogenic ^{129}I is retained in the upper 10 cm of the soil column. Data from 123 soil cores from nonagricultural lands on the SRP give an average volumetric soil density of 1.2 g/cm³ for the top 10 cm (Beasley and others, 1998). Thus, the calculated concentration of ^{129}I in SRP soils contaminated by integrated global fallout is approximately 0.8 aCi/g (2×10^7 atoms/g). This is about two orders of magnitude larger than the concentration estimated to result from the steady-state deposition of naturally produced ^{129}I in the atmosphere to near-surface SRP soils, even when considering that this naturally produced ^{129}I is likely to be concentrated in the upper few centimeters of undisturbed soils. This calculated concentration does include regionally derived ^{129}I input from testing at the Nevada Test Site (NTS), however, at present this input cannot be quantified precisely.

Nuclear-Fuel Reprocessing Facilities Releases

Over the operating history of the INTEC, an estimated 1.1×10^{26} atoms (4 Ci) of ^{129}I have been released to the atmosphere (Mann and others, 1988). A comparable release has been estimated for nuclear-fuel reprocessing facilities at the Savannah River site near Aiken, South Carolina (4.7 Ci, or 1.2×10^{26} atoms; Kantelo and others, 1982). Approximately 51 Ci (1.4×10^{27} atoms) of ^{129}I was present in nuclear fuels from the Hanford plutonium production reactors that were processed from 1944 to 1986 (Robkin and Shleien, 1995). Similarly, large amounts of ^{129}I have been released from the nuclear-fuel reprocessing facilities at Sellafield, United Kingdom, and Cap de la Hague, France (180 Ci, or 4.8×10^{27} atoms) to the ocean alone; (Raisbeck and others, 1994). Atmospheric releases of ^{129}I from these two European facilities are thought to have been transported into the Arctic Ocean Basin as far north as Mould Bay on Prince Patrick Island, Canada (Kilius and others, 1995), and, hence, also could contribute to fallout at the INEEL.

Depth of sampling is important in the comparisons discussed in the preceding paragraphs; therefore, the comparison made here is qualitative at best. Concentrations of ^{129}I in the upper 5 cm of soil (where ^{129}I is often efficiently retained) are expected to be higher than concentrations in the upper 10 cm of the soil column because of greater dilution with ^{129}I -free soil as depth of sampling increases. However, the pronounced differences in soil ^{129}I concentrations measured near and at great distances from nuclear-fuel reprocessing facilities confirm their importance in influencing environmental inventories of ^{129}I over regional, continental, and global scales. The concentration of ^{129}I in soil within 2 km of the INTEC is as large as 10,400 aCi/g (2.8×10^{11} atoms/g); at Island Park, Idaho (200 km northeast of the INEEL; fig. 1), the concentration of ^{129}I in soil is 48 aCi/g (1.3×10^9 atoms/g). This concentration is more than two orders of magnitude smaller than that in soils close to the INTEC but nearly 2 orders of magnitude larger than that predicted as a result of global weapons-tests fallout (about 1 aCi/g, or 2×10^7 atoms/g, Gove and others, 1994). Brauer (1974)

recorded ^{129}I concentrations in surface soil in Washington, Idaho, and Montana ranging from 2 aCi/g to 8 aCi/g (5×10^7 and 2.8×10^8 atoms/g), respectively, concentrations similar to those expected from global fallout. However, concentrations of ^{129}I in soil within 200 km of the Hanford site were larger than those reported by Brauer (1974) by as much as two orders of magnitude as a result of nuclear-fuel reprocessing releases. At Richland, Washington, near Hanford, concentrations of ^{129}I in surface soil were as large as 1,600 aCi/g (4×10^{10} atoms/g) (Brauer and Strebin, 1982).

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the work on an early draft of this research by Larry J. Mann (USGS, retired) and Tom M. Beasley (DOE, retired). Additionally, thanks are due to David Frederick, State of Idaho INEEL Oversight Program and F.P. Brauer, formerly with the Pacific Northwest National Laboratory, Richland Washington for their review of an early draft of this work. We also acknowledge the earlier studies undertaken to establish a background ^{129}I concentration in water from the eastern SRP and the extensive sample collection and sample processing efforts (Mann and Beasley, 1994a, 1994b).

The present report was reviewed by Dr. June Fabryka-Martin with Los Alamos National Laboratory, New Mexico, and Ms. Kristi Moser-McIntire, State of Idaho INEEL Oversight Program. We thank them for their constructive comments that improved the technical content and clarity of this report.

METHODS AND QUALITY ASSURANCE

The 34 samples from the 32 sites and the 2 QA/QC samples included in this study were collected, processed, and analyzed in the same manner as the 16 samples previously described by Mann and Beasley (1994a). Water samples from streams were collected downstream from riffles to ensure that the surface water was well homogenized. At sites where the stream was shallow and could be waded, samples were obtained from at

least three locations across the stream and composited into a single sample for field processing. For deeper streams, water was collected as far as possible from each bank where the water was at least 1 m deep and well mixed. Following collection, the two samples were composited into a single sample.

Field processing consisted of filtering the water through a disposable, 0.45- micrometer filter cartridge that had been pre-rinsed with at least 1 L of deionized water and 1 L of water from the field site. Filtration was necessary to remove particulate matter which could affect the laboratory preparation of the silver-iodide targets used in the AMS measurements of ^{129}I . In addition to the 34 primary samples, 2 blind replicate samples were submitted to the laboratory for analysis (tables 2 and 3).

Blind replicate QA/QC samples were analyzed from the Portneuf River near Tyhee and from the Unnamed Spring No. 2 above Shoshone Powerplant near Twin Falls (fig. 1 and table 2). Relative percent differences for these replicate pairs were 15 and 40, respectively. Six samples taken from USGS archives are included in table 2. All the archived samples were collected in 1992, and among them were samples from Box Canyon Springs near Wendell and Briggs Creek near Buhl, which were also sampled in 1994. Results for the archived 1992 samples were compared with results from the samples collected in 1994 at Box Canyon Springs near Wendell and Briggs Creek near Buhl. Relative percent differences were 27 and 29; ^{129}I concentrations in both 1992 samples were smaller than in the 1994 samples. In addition to natural heterogeneity between replicate water samples, factors contributing to differences in analytical results include incomplete equilibration of iodide carrier with ^{129}I in the sample and cross contamination of the prepared sample during processing or in the ion source of the accelerator during the sputtering process (Mann and Beasley, 1994a; David Elmore, Purdue University, written commun., 2002). Impact to the final analytical results from incomplete equilibration and cross contamination are lessened by the standard proto-

col of repeating measurements that are averaged to calculate the final ratio of $^{129}\text{I}/^{127}\text{I}$ (David Elmore, Purdue University, written commun., 2002).

RESULTS OF IODINE-129 ANALYSES

The present study combines 52 results from 48 sites: 20 samples and 2 QA/QC samples collected in 1993-94, 6 samples from USGS archives that were collected in 1992, 8 samples collected in 1993, and 16 samples collected in 1992. Subsets of this analytical data set were identified for calculation of summary statistics to evaluate possible trends with respect to sample type and geographic location and to quantify uncertainties for subpopulations. Site locations are shown in figure 1. Concentrations of ^{129}I for these samples are listed in tables 1-3. Sample sites from the study by Mann and Beasley (1994a) are listed in table 1 with recalculated results for this study. Results for ^{129}I in water samples from streams and springs that were collected for this reevaluation, or that were selected from the USGS sample archives are shown in table 2. Results for ^{129}I in ground-water samples from selected sites in the USGS sample archives or samples collected in 1993 are shown in table 3. Archived ground-water samples listed in table 3 are from five U.S. Bureau of Land Management-owned stock wells sampled in 1993 for selected radiochemical and water chemistry to address public concerns about the impacts of INEEL activities on ground water (Bartholomay and others, 2001). The results for the blind replicates (QA/QC samples) from the Portneuf River near Tyhee and for the Unnamed Spring No. 2 above Shoshone Power Plant near Twin Falls (table 2) were not included in the calculation of the background ^{129}I concentration. The results for the 1994 primary and 1992 archived samples for Box Canyon Springs near Wendell and Briggs Creek near Buhl (table 2) were included in the summary statistics.

Sample sites were categorized by a combination of their relative location (eastern SRP or tributary valleys) and by the type of site (surface water, or water from springs and wells). Eastern SRP sites are wells, springs, and surface-water sites within the boundaries of the plain (fig. 1). The

Snake River at King Hill (the outflow point for the eastern SRP and tributary basins) was included in this subset. Wells that were not completed in the eastern SRP aquifer, and springs or surface water sites whose sources or drainage basins lie outside the eastern SRP, were categorized as tributary sites. Although the sampling sites Henrys Fork at St. Anthony and the Portneuf River near Tyhee are located within the boundaries of the eastern SRP, most of their drainage basins lie outside of the plain, so these two sites were categorized as tributary sites. The category of ground-water sites includes wells completed in, and springs that discharge from, the eastern SRP aquifer. Excluded from this category are samples from Lidy Hot Springs and the City of Arco well as these sites are tributary to the SRP.

Iodine-129 concentrations for these 50 samples (minus the 2 replicate samples) ranged from 0.29 ± 0.18 aCi/L to 30.3 ± 6.2 aCi/L. The largest ^{129}I concentrations were measured in streams tributary to the SRP: Big Wood River near Bellevue (30.3 ± 6.2 aCi/L), Silver Creek near Picabo (24.6 ± 0.7 aCi/L), Beaver Creek at Spencer (18.0 ± 1.3 aCi/L), Henrys Fork near Island Park (12.7 ± 0.5 aCi/L), and Medicine Lodge Creek near Small (10.0 ± 0.3 aCi/L). Coffee Point (CCC-3), a well completed in the SRP aquifer, contained the same concentration as Medicine Lodge Creek near Small (10.0 ± 0.4 aCi/L) (table 2). The smallest ^{129}I concentrations were measured in a sample from the Teton River near St. Anthony (0.49 ± 0.10 aCi/L) and water samples from wells MV-59 (0.29 ± 0.18 aCi/L) and MV-04 (0.44 ± 0.19 aCi/L) (tables 2 and 3, respectively). The Teton River near St. Anthony is tributary to the plain, and wells MV-59 and MV-04 are completed in the eastern SRP aquifer south of the INEEL (fig. 1).

The log-normal distribution is common with variables having a natural lower or upper limit and is prevalent in geochemical data (Rock, 1988). A log-normal distribution can be identified by testing for normality on the natural logs of the data. A natural log transform was applied to the data to test for normality using the Shapiro-Wilk W test (Royston, 1992). Because the W test ($W = 0.9322$, $p = 0.0067$) shows that the data were not normally

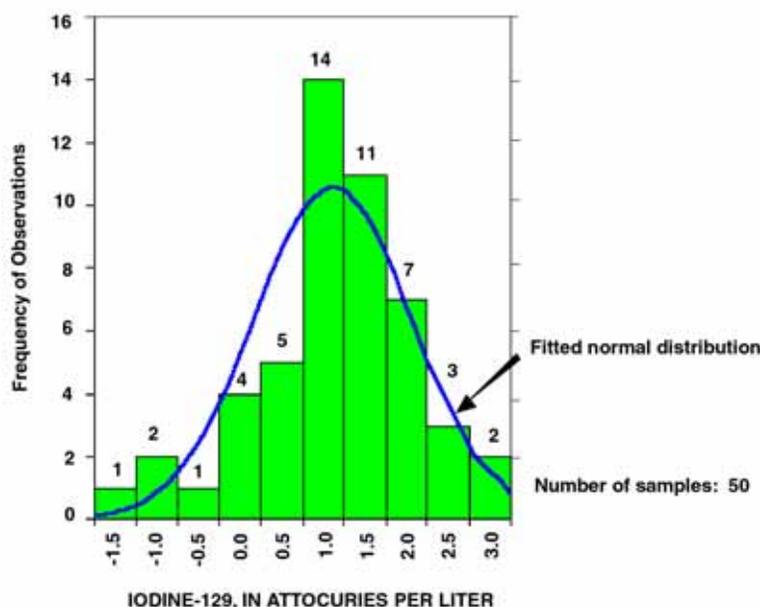


Figure 2. Histogram of the natural log transform (Ln) of iodine-129 concentrations in spring water, ground water, and surface water, eastern Snake River Plain, Idaho, 1992–94.

distributed (and more nearly resembled a log-normal distribution, fig. 2) nonparametric statistics were used to analyze the data. The median (the 50th-percentile value) is a resistant statistical measure only minimally affected by outliers (Helsel and Hirsch, 1992) and best reflects the central tendency of these data. A corresponding measure of the spread of the data is the interquartile range (the difference in value between the 25th and 75th percentiles).

Summary statistics that include maximum, minimum, median and associated 95-percent confidence interval, the interquartile range, and the 95th-percentile values for subpopulations based on sample locations are presented in table 4. The 95th-percentile values for each subset and associated 95-percent confidence intervals were calculated using nonparametric statistical methods (Helsel and Hirsch, 1992). The median concentration for sites tributary to the plain (8.4 aCi/L and the 95-percent confidence interval is 2.6 to 10.0 aCi/L) is larger than the median concentration for sites on the plain (3.4 aCi/L and the 95-percent confidence

interval is 3.0 to 4.4 aCi/L). The median concentration for surface-water sites (8.2 aCi/L and the confidence interval is 5.7 to 10.0 aCi/L) is larger than the median for the ground-water sites (3.3 aCi/L and the confidence interval is 2.5 to 4.1 aCi/L, table 4). The differences between ground-water sites and surface-water sites are probably a result of evapotranspiration which concentrates the ¹²⁹I in surface water and in recharge to ground water from irrigation compared to the ground-water concentrations.

The box-whisker plots in figure 3 illustrate the absolute and relative values of the median, the confidence interval about the median, the interquartile range, and the potential outliers for the subsets of data evaluated. Little variation is evident in ¹²⁹I concentrations from the spring sites (fig. 3). Although the range of concentrations is larger for wells, the medians do not differ statistically for these data sets. Three possible outliers are evident from the diagram for surface-water sites. The larger median concentration for surface-water sites is also clearly apparent.

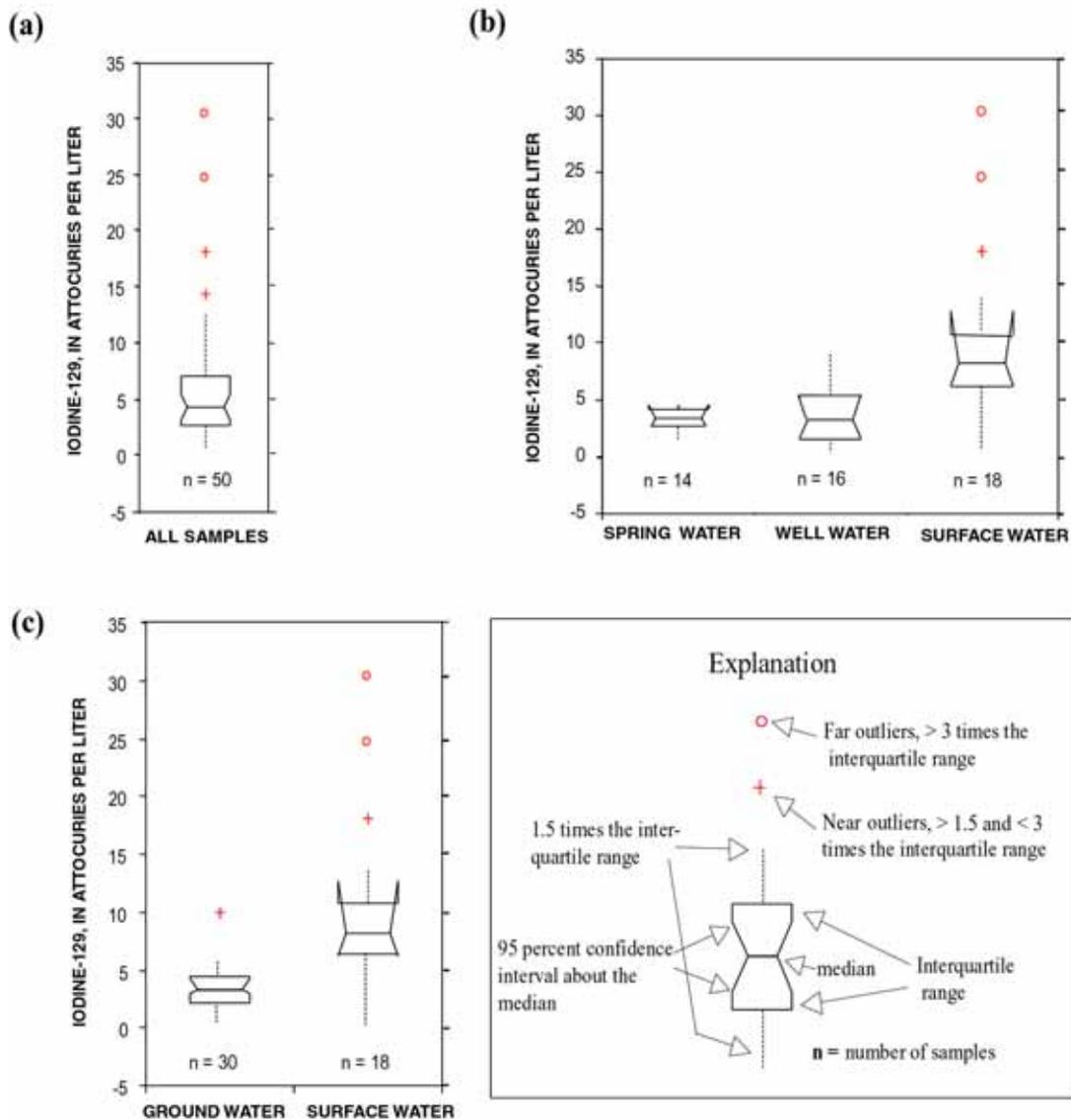


Figure 3. Iodine-129 concentrations in spring water, ground water, and surface water, eastern Snake River Plain, Idaho, 1992-94.

(a) All samples. (b) Samples distinguished by source; spring water, ground water (water from wells completed in the Snake River Plain aquifer), and surface water. (c) Ground water (includes spring water and water from wells completed in the Snake River Plain aquifer) and surface-water samples. Samples from the City of Arco well and Lidy Hot Springs are excluded from (b) and (c), see text for explanation.

In this report, review of the ^{129}I results from all 50 samples (minus the 2 replicate samples) suggests that the background concentration for the SRP should be estimated using only data for samples collected from wells completed in the aquifer and springs discharging from the aquifer that are unaffected by disposal practices at the INEEL, and that surface-water samples should be excluded. This concentration at the 95th-percentile value is 5.4 aCi/L, and the 95-percent confidence interval about that value ranges from 5.2 aCi/L to 10.0 aCi/L (table 4). Mann and Beasley (1994a) determined that results for 16 ground-water, surface-water and spring-water sites sampled in 1992 were normally distributed, and constructed a parametric, 99-percent, one-tailed confidence interval to establish an upper bound of 8.2 aCi/L for the combined contributions of naturally produced ^{129}I and ^{129}I from global fallout from atmospheric weapons tests.

One ground-water sample was identified as an outlier in the statistical analysis. The result from 1993 for water from the Coffee Point well (CCC-3; 10.0 ± 0.4 aCi/L) defines the upper bound to the 95-percent confidence interval for the 95-percent value for ground water from the SRP aquifer in figure 3. Followup sampling in 1998 by the State of Idaho INEEL Oversight Program at this same site yielded a result of 0.16 ± 0.013 aCi/L (L. F. Hall, unpubl. data, 2002), which suggests that the 1993 Coffee Point result may represent a transient event and, thus, may not be representative of background concentrations or may reflect sample contamination. Because nonparametric statistical methods were utilized in this reevaluation, the median and the 95-percent value for SRP aquifer wells and springs would not be changed significantly (from 3.3 to 3.2 aCi/L for the median and 5.4 to 5.3 for the 95th-percentile value) by excluding the 1993 Coffee Point result from the calculations. However, excluding this result from the calculations would change the confidence interval about the median (from 3.1 to 4.4 aCi/L, to 2.5 to 3.9 aCi/L), and the confidence interval about the 95th-percentile value (from 5.3 to 10.1 aCi/L, to 4.6 to 5.3 aCi/L).

The principal sources of ^{129}I in wells and springs on the eastern SRP for those locations that can be reasonably assumed to be uncompromised by disposal of low-level radioactive waste from the INEEL are infiltration of recharge water containing ^{129}I from anthropogenic sources other than the INEEL (global fallout) and natural production in the atmosphere and the lithosphere. *In situ* production in the aquifer matrix is considered to be a nonexistent or minor source of ^{129}I .

Cecil and others (2000) presented the results of calculations for SRP rock samples and showed that the *in situ* ^{36}Cl production within the rock matrix is negligible and is on the same order of magnitude as natural atmospheric production, which suggests that the ^{129}I *in situ* production would be insignificant as well. *In situ* production is considered small, not only because of the relatively small U content and young age of the SRP basalt, as discussed earlier, but also because the residence time of water in the eastern SRP aquifer is estimated to be generally less than 300 years (Ackerman, 1995; Cecil, 2000), a time far too short to allow significant ^{129}I buildup from the spontaneous fission of U isotopes. Lindholm (1988) estimated that in 1980, the eastern SRP received 8.3×10^9 m³ of recharge and, of this total, 60 percent (5×10^9 m³) was attributable to irrigation return and 20 percent (1.7×10^9 m³) was a result of tributary flow. If these volumes are representative of recharge over the past two decades, and given that nearly one-third of the entire SRP area is presently under irrigation (Lindholm, 1988), additional ^{129}I could move from the landscape to surface and ground water. The efficiency of this removal process, however, has not been determined.

On the basis of reevaluation of the sample results presented in this report, it was concluded that the maximum background concentration for ^{129}I in ground water from the eastern SRP at the 95th-percentile value is 5.4 aCi/L, and the nonparametric confidence interval is 5.2 to 10.0 aCi/L. Concentrations above this level may be a result of evapotranspiration processes, resuspension and concentration of fallout from anthropogenic sources, or contamination from wastewater disposal at the INEEL.

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Table 1. Concentrations of iodine-129 in water from selected sites on or tributary to the eastern Snake River Plain, Idaho (recalculated and modified from Mann and Beasley, 1994a).

Site name (figure 1)	Site identification number	Date sampled (m/d/y) ^a	¹²⁹ I ^b		Sample type
			(aCi/L) ^c	atoms/L ^d (x 10 ⁶)	
Springs and streams^e					
Birch Creek at Blue Dome	13117020	4/6/92	2.1±0.4	56.8±9.44	Surface water
Little Lost River near Howe	13119000	4/6/92	7.9±0.8	209±21.2	Surface water
Big Lost River at Mackay	13127700	4/6/92	2.6±0.4	69.8±10.6	Surface water
Crystal Spring near Buhl	13093396	4/6/92	3.0±0.4	78.6±10.9	Spring water
Banbury Springs near Buhl	13095300	3/2/92	3.0±0.5	78.1±12.0	Spring water
Thousand Springs near Hagerman	13132800	3/3/92	1.5±0.4	39.7±10.6	Spring water
Birch Creek Spring near Hagerman	13135100	3/3/92	5.2±0.4	137±11.5	Spring water
Wells					
Atomic City well	432638112484101	1/22/92	1.1±0.4	28.4±11.6	Ground water
City of Arco well	433758113181701	6/2/92	1.0±0.4	26.1±9.73	Ground water
P&W 2	435419112453101	4/8/92	5.3±0.5	141±12.5	Ground water
Well 4 (USGS-004)	434657112282201	4/24/92	5.3±0.6	141±15.6	Ground water
Well 19 (USGS-019)	434426112575701	4/8/92	1.6±0.4	42.9±10.3	Ground water
Well 26 (USGS-026)	435212112394001	4/21/92	3.9±0.5	102±12.5	Ground water
Well 97 (USGS-097)	433807112551501	3/16/92	4.3±0.5	114±13.5	Ground water
Well 100 (USGS-100)	433503112400701	3/27/92	2.0±0.4	52.1±10.3	Ground water
Well 110 (USGS-110)	432717112501501	1/22/92	3.1±0.4	82.6±11.5	Ground water

a. Date format: month/day/year.

b. Uncertainties at 1 sample standard deviation.

c. Attocuries per liter.

d. Atoms per liter.

e. Springs and streams listed in order of downstream ground-water flow.

Note: iodide concentrations were not measured at the time of sampling; a concentration of 0.001 mg/L was assumed for the purpose of calculating ¹²⁹I results.

Table 2. Field measurements and concentrations of iodine-129 in surface water and springs at selected sites on or tributary to the eastern Snake River Plain, Idaho (in order of downstream flow).

Site	Site identification number	Date sampled (m/d/y) ^a	Discharge (m ³ /s) ^b	Water temp ^c	pH	Specific conductance ^d	Dissolved iodide ^e	¹²⁹ I ^f		Sample type
								(aCi/L) ^g	atoms/L ^h (x 10 ⁶)	
Snake River near Irwin	13032500	3/2/94	41.6	2.5	7.9	370	0.002	7.0±0.3	184±7.49	Surface water
Snake River at Heise Bridge	433847111420000	3/2/94	57.2	6.0	7.6	482	0.003	7.1±0.4	188±9.42	Surface water
Henrys Fork near Island Park	13042500	3/3/94	12.5	5.0	7.8	159	0.001	12.7±0.5	336±12.2	Surface water
Henrys Fork at St. Anthony	13050500	3/3/94	52.1	5.5	8.5	170	0.001	8.6±0.4	226±9.70	Surface water
Teton River near St. Anthony	13055000	3/2/94	12.1	6.5	7.8	340	0.001	0.49±0.10	12.9±2.56	Surface water
Snake River near Shelley	13060000	3/1/94	89.8	4.5	8.1	319	0.002	9.2±0.5	243±13.1	Surface water
Blackfoot River near Shelley	13066000	3/1/94	2.3	5.5	8.3	450	0.003	14.3±0.6	379±17.2	Surface water
Portneuf River near Tyhee	13075910	3/4/94	15.2	8.5	7.7	650	0.004	8.4±0.3	222±7.77	Surface water
								9.8±0.7	259±19.7	Blind replicate
Snake River at Neeley	13077000	3/4/94	64.6	8.0	8.5	590	0.003	6.2±0.3	164±7.22	Surface water
Snake River at Milner	13088000	3/4/94	6.7	6.5	8.5	590	0.004	7.5±0.4	198±9.43	Surface water
Devils Washbowl Spring near Kimberly	13089600	3/4/92 ⁱ	0.4	14.0	8.3	638	j	3.4±0.2	90.5±5.78	Spring water
Devils Corral Spring near Kimberly	13090100	3/4/92 ⁱ	1.1	15.0	8.0	630	j	2.5±0.2	67.4±6.39	Spring water
Unnamed Spring No 2 above Shoshone Powerplant near Twin Falls	13090350	3/4/92 ⁱ	0.1	14.0	8.5	425	j	4.1±0.3	108±9.11	Spring water
								2.7±0.2	71.5±4.98	Blind replicate
Blue Lakes outlet near Twin Falls	13091500	3/9/94	4.1	16.0	8.0	630	0.002	3.4±0.4	91.0±11.5	Spring water
Warm Creek near Twin Falls	13091700	3/9/94	0.6	15.0	7.8	650	0.002	4.6±0.5	121±12.0	Spring water
Briggs Creek near Buhl	13095200	3/7/94	3.0	14.5	7.8	490	0.002	4.4±0.2	115±5.84	Spring water
		3/2/92 ⁱ	2.8	14.0	7.9	451	j	3.3±0.5	86.1±14.4	Spring water
Box Canyon Springs near Wendell	13095500	3/8/94	9.5	14.5	7.6	400	0.002	4.6±0.7	121±19.4	Spring water
		3/2/92 ⁱ	9.0	14.0	7.9	408	j	3.5±0.4	92.8±11.4	Spring water
Beaver Creek at Spencer	13113000	3/3/94	0.4	0.5	8.2	410	0.002	18.0±1.3	477±33.7	Surface water
Medicine Lodge Creek near Small	13116500	3/3/94	1.5	6.0	8.4	430	0.001	10.0±0.3	265±6.94	Surface water
Lidy Hot Springs	440832112331001	3/14/94	j	47.0	7.3	680	0.004	3.7±0.3	98.9±7.77	Spring water
Sand Springs near Hagerman	13132600	3/2/92 ⁱ	2.1	14.0	7.8	380	j	3.2±0.4	84.2±9.70	Spring water
Big Wood River near Bellevue	13141000	3/5/94	1.4	5.0	7.2	380	0.003	30.3±6.2	800±165	Surface water
Silver Creek near Picabo	13150430	3/5/94	6.3	7.0	7.3	420	0.002	24.6±0.7	650±18.0	Surface water
Snake River at King Hill	13154500	3/5/94	248.7	11.0	7.9	510	0.003	5.7±0.3	150±8.87	Surface water

a. Date format: month/day/year.

b. Cubic meters per second.

c. Degrees Celsius.

d. Microsiemens per centimeter at 25°C.

e. Milligrams per liter.

f. Uncertainties at 1 sample standard deviation.

g. Attocuries per liter.

h. Atoms per liter.

i. Archived sample.

j. Not measured, assumed to be 0.001 mg/L for calculation of the iodine-129 concentration.

Table 3. Field measurements and concentrations of iodine-129 in water from selected wells downgradient from the Idaho National Engineering and Environmental Laboratory, Idaho.

Well	Site identification number	Date sampled (m/d/y) ^a	Water temp ^b	pH	Specific conductance ^c	¹²⁹ I ^d		Sample type
						(aCi/L) ^e	atoms/L ^f (x 10 ⁶)	
Coffee Point (CCC-3)	430911112585401	6/21/93 ^g	15.0	8.1	310	10.0±0.4	265±10.2	Ground water
Crossroads	432128113092701	6/22/93 ^g	11.0	8.2	337	3.2±0.2	84.2±4.91	Ground water
Fingers Butte	432424113165301	6/22/93 ^g	15.0	8.3	306	5.5±0.2	146±6.04	Ground water
Grazing-2	431553112492001	6/21/93 ^g	15.0	8.1	290	1.5±0.1	40.7±3.31	Ground water
Houghland	431439113071401	6/22/93 ^g	16.0	8.1	327	4.5±0.2	120±5.70	Ground water
MV-04	424658113414601	8/2/93	12.5	7.8	661	0.44±0.19	11.7±4.90	Ground water
MV-33	425455114160101	8/2/93	16.0	7.9	297	1.7±0.2	44.6±5.63	Ground water
MV-59	430805113102401	8/10/93	15.0	8.0	306	0.29±0.18	7.72±4.63	Ground water

a. Date format: month/day/year;

b. Degrees Celsius.

c. Microsiemens per centimeter at 25°C.

d. Uncertainties at 1 sample standard deviation.

e. Attocuries per liter.

f. Atoms per liter.

g. Archived sample.

Note: iodide concentrations were not measured at the time of sampling; a concentration of 0.001 milligrams per liter (mg/L) was assumed for the purpose of calculating ¹²⁹I results.

Table 4. Summary statistics for selected iodine-129 data sets from sites on or tributary to the eastern Snake River Plain, Idaho.

Data set	Number of samples	Concentration (attocuries per liter) ^a						
		Maximum	Minimum	Median	95-percent confidence interval about the median	Inter-quartile range	95th percentile value	Approximate 95 percent confidence interval for the 95 th percentile value
Comparison of data sets from different studies								
Background samples, 1992 ^b	16	7.9±0.8	1.0±0.4	3.0	1.6–3.9	2.7	5.8	5.3–7.9
Background samples, 1993-94, and archived samples	34	30.3±6.2	0.29±0.18	4.6	3.4–7.0	5.3	20.0	12.7–30.3
Combined data sets, 1992 and 1993-94 with archived samples	50	30.3±6.2	0.29±0.18	4.2	3.2–5.2	4.4	16.2	10.0–30.3
Comparison of data from sites on the SRP with data from sites tributary to the SRP								
Sites on the eastern Snake River Plain	33	10.0±0.4	0.29±0.18	3.4	3.0–4.4	2.5	6.7	5.5–10.0
Sites in areas tributary to eastern Snake River Plain	17	30.3±6.2	0.49±0.10	8.4	2.6–10.0	9.1	25.4	18.0–30.3
Comparison of surface-water data with data from wells and springs								
Surface-water data	18	30.3±6.2	0.49±0.10	8.2	5.7–10.0	5.4	25.2	18.0–30.3
Ground water data (wells and springs from the Snake River Plain aquifer)	30 ^c	10.0±0.4	0.29±0.18	3.3	2.5–4.1	2.6	5.4	5.2–10.0

a. Uncertainties at 1 sample standard deviation.

b. Data from Mann and Beasley (1994a).

c. Samples from Lidy Hot Springs and the City of Arco well are excluded.

This summary does not include the two replicate samples discussed in the text and listed in table 2.